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Pulsed Neutron Research for Nuclear Safeguards

Program Status Report

January-March, 1967

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NUCLEAR SAFEGUARDS RESEARCH SERIES

G. Robert Keepin, Editor

This LA...MS report, the first in the LASL Nuclear Safeguards Research Series, presents the status of the nuclear safeguards research program at Los Alamos.

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PULSED NEUTRON RESEARCH FOR NUCLEAR SAFEGUARDS

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NUCLEAR SAFEGUARDS RESEARCH PROGRAM AT LOS ALAMOS, BACKGROUND AND PURPOSE

The Los Alamos Scientific Laboratory has undertaken a new research and development program in the technology of inspection, surveillance and control of fissionable material and fission devices. The urgency and importance attached to this new technical field of nuclear safeguards has been repeatedly stressed by national leaders at the highest levels in both the legislative and executive branches of government (including specifically, the President of the United States).

In view of the growing necessity for more comprehensive and effective domestic and international control of nuclear energy and material, the field of nuclear safeguards is destined to assume major importance in the era of widespread nuclear power proliferation (and its inevitable concomitant, fissile materials proliferation) which lies just ahead. Based on extensive experience and leadership in technical fields closely related to safeguards technology, LASL is uniquely qualified to provide national leadership in this new technical field of nuclear energy safeguards and control.

The technical aspects of nuclear safeguards divide logically into two categories. These are: (1) the detection of nuclear detonations, in which the Laboratory is already contributing via Vela Hotel, Vela Sierra, and Long Range Detection Programs, and (2) the technology of inspection, surveillance and control of fissionable material. To provide urgently needed national leadership in the technology of nuclear safeguards, a new Pulsed Neutron Research Group (N-6) has been formed at Los Alamos, with the specific mission of developing promising new approaches to the detection, identification and analysis of fissionable materials.

TECHNICAL PROGRAM OF PULSED NEUTRON RESEARCH GROUP

The technical program of the LASL Pulsed Neutron Research Group (N-6) includes measurements of the detailed kinetics and energy characteristics of delayed fission neutrons, as well as fission product gamma and β emitters. Both basic research into delayed neutron properties and the development of practical systems utilizing delayed neutron response techniques for detection, identification and analysis (DIA) of fissionable material are necessary phases of this technical program. Some of the required basic measurements to be carried out by Group N 6 are:

a. Absolute delayed neutron yield at high incident neutron energy. (Existing yield and abundance discrepancies at high energies will be fully and unequivocally resolved.)

b. Delayed neutron group abundances and decay constants for 14 MeV-neutron-induced fission of all major fission species (these measurements are required for optimum exploitation of delayed neutron response methods).

c. Energy spectra of individual delayed neutron groups from the major fissioning species. Neutron spectrometer development. (Investigation of increased isotope discrimination factors using energy-sensitive neutron detectors.)

d. Extensive studies of the neutron-rich fission products, particularly delayed neutron (and later delayed gamma) emitters, with a view toward advanced DIA applications.

As nearly coincident as possible with the required basic measurements listed above, Group N-6 will apply delayed neutron and kinetic response techniques to the development of practical DIA systems. This phase of the technical program will begin with experimental investigations of the new $\bar{R}_f$ and $\bar{S}_f$ methods (cf. G. R. Keepin, U. S. Patent Disclosure, AEC No. S-34-395) for non-destructive detection, identification and analysis of fissionable materials. These methods are based on characteristic differences in relative group yields and energy spectra of delayed neutrons from the various fission species. In essence, the methods consist of detailed analyses (in time and, as a further refinement, in energy) of the measured delayed neutron activity from an unknown system following pulsed neutron irradiation.

The experimental techniques involved are rapid, non-destructive, and relatively simple and inexpensive. No absolute calibrations (of source, detectors, fission rates, etc.) are required, and all measurements are made in a time domain which is completely free of perturbations due to prompt neutron higher modes, diffusion and thermalization effects. Further important advantages are "environment insensitivity" (in the presence of high radiation fields, inert materials, etc.) and the high "penetrability" of fast neutrons through high density bulk materials.

The high sensitivity of the delayed neutron response methods for discrimination between different fissionable isotopes is indicated by the measurable "isotope discrimination factors" shown in Table I. It is clear that the $\overline{R}_f$ and/or $\overline{S}_f$ methods should be well suited, for instance, to the assay of Pu^{239} (or U^{233}) in power reactor fuel elements or breeder blanket material, or to rapid, non-destructive evaluation of burnup in irradiated fuel. Similarly, the large effective discrimination factor between U^{235} and U^{238} enables direct determination of isotopic abundances in enriched uranium fuel samples or in U^{235}-U^{238} composite systems or devices.

TABLE I

EFFECTIVE ISOTOPE DISCRIMINATION FACTORS

(R_f and S_f Measurements with Flat Response Detector)

Fission Isotopes	Effective Discrimination Factor (R_{f-} and R_{f+})	Effective Discrimination Factor (R_{f-} and $S_{f+/\Delta}$)
U^{238}, U^{235}	3.30	3.80
U^{238}, Pu^{239}	4.60	5.20
U^{238}, U^{233}	7.30	10.20
U^{235}, Pu^{239}	1.38	1.33
U^{235}, U^{233}	2.15	2.60
Pu^{239}, U^{233}	1.58	1.94

Based on calculated end point energies for the delayed neutron emission spectra from identified fission product precursors, considerably increased discrimination factors between the various fissioning species can be further realized by the use of energy-sensitive neutron detectors.

The development of practical DIA systems will include studies of a variety of differential shielding (transmission) effects in representative types of systems, the development of high-efficiency neutron spectrometers and optimization of detectors to achieve maximum isotope discrimination. Empirical approaches to DIA will also include measuring and cataloging delayed neutron response "signatures" for various types of systems, determining the ultimate sensitivity of DIA systems in identifying "active" (fissile) versus "safe" (fertile) fissionable materials, and identifying and analyzing unknown or clandestine systems. Advanced experimental and analysis techniques will be pursued through multi-dimensional multichannel time and energy analysis, development of high intensity pulsed sources, and the extension of delayed neutron response methods to the determination of absolute amounts of fissionable materials present in unknown systems (using absolute source and detector calibration techniques). Finally, hardware concepts will be implemented to develop an integrated source-detector-analyzer system with demonstrated practical performance for various types of inspection and surveillance applications.

PULSED COCKCROFT-WALTON ACCELERATOR AND AUXILIARY EQUIPMENT

The principal research tool of Group N-6 at present is a 350 KV pulsed Cockcroft-Walton accelerator which was recently installed in its present location at TA-18, Los Alamos. This versatile pulsed neutron source is operated in both the direct current and pulsed modes with a total neutron source strength in excess of 10^{11} neutrons/sec using the D(T,n) He^3 reaction. In the pulsed mode, the accelerator can produce neutron pulses of any desired width down to the nanosecond region (required for prompt neutron measurements on fast metal systems) with corresponding pulse repetition rates variable up to the megacycle range. A 5 KV beam deflection system was recently developed for post-acceleration pulsing to supplement the existing pre-acceleration pulsing capability in the high voltage head.

Because of its versatility and uniqueness at LASL as a flexible, intense pulsed neutron source, operating time on the N-6 accelerator is in considerable demand by other LASL technical groups. In addition to research use by N-6, during the first quarter of 1967 the accelerator was used by technical groups in the Test Division (J), the Physics Division (P), the Explosives Technology Division (GMX) and the Weapons Division (W).

A 50 channel high-speed recording time delay analyzer, designed specifically for use with the N-6 accelerator, is used for data acquisition in the microsecond to the seconds time range. For pulsed neutron measurements in the submicrosecond range (e.g. prompt neutron decay experiments), a precision time-to-pulse height converter is used in conjunction with multichannel pulse height analyzers.

ABSOLUTE DELAYED NEUTRON YIELD MEASUREMENTS

As part of the program of basic data required for the development of practical DIA systems, the absolute yield of delayed neutrons from high energy fission of the major fission species are currently being measured using the pulsed CW accelerator as the neutron source. The general experimental arrangement is shown in Figure 1. The beam accel-

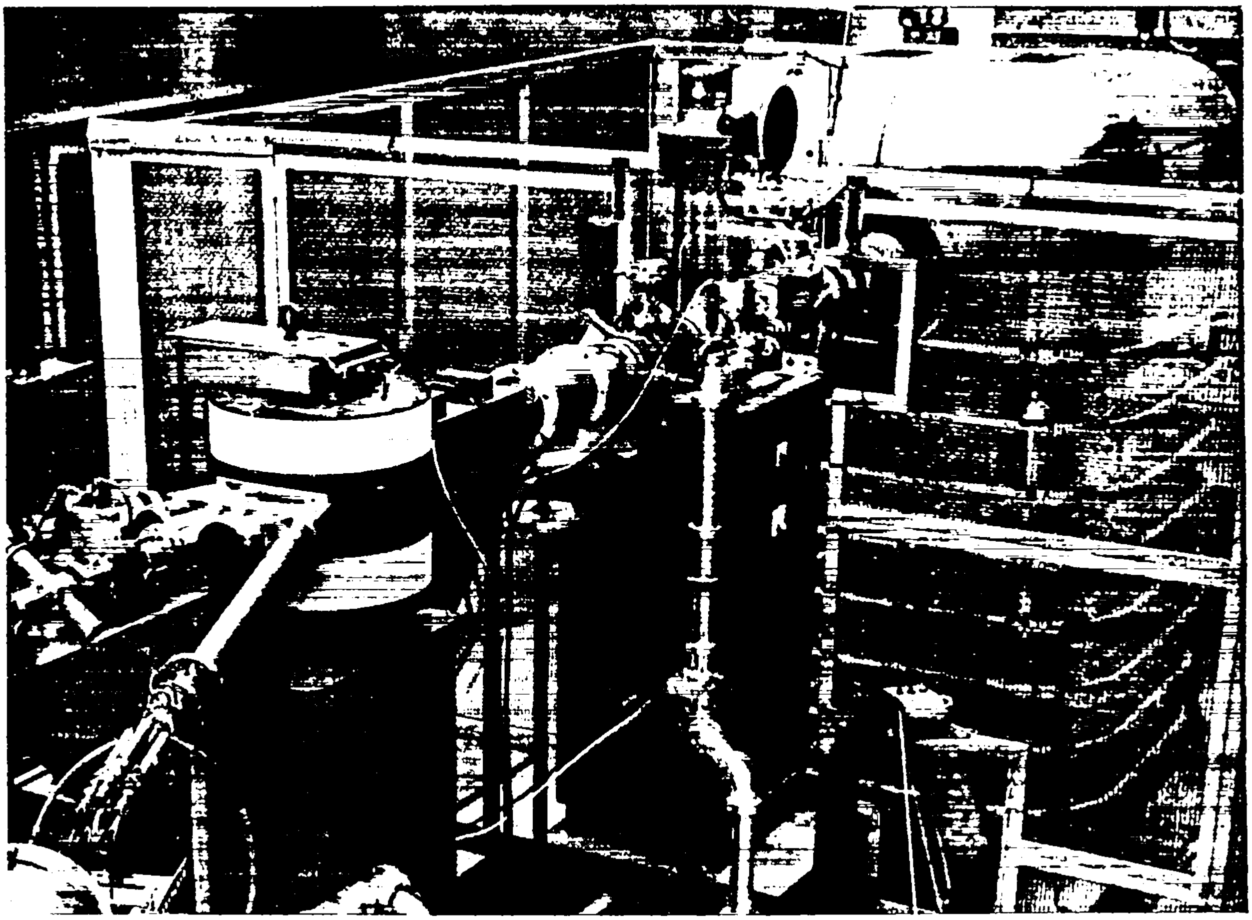

Fig. 1. Experimental arrangement for absolute delayed neutron yield measurements.

eration tube can be seen extending from the high voltage deck (containing ion source, pre-acceleration pulsing equipment, etc.) at the upper right to the vacuum cabinet, beam deflection magnet, and thence through the 25°-left beam port to the target and multiple detector configuration at the lower left.

Absolute delayed neutron yield measurements at high energies are of particular interest since a basic discrepancy persists between experimental and theoretically predicted yields at high energies -- notably at 14 MeV. Long standing experimental data (from both the USA and the USSR) have indicated a strong increase -- roughly a doubling -- in delayed neutron yield as one progresses from low energy fission to 14 MeV fission. This contrasts sharply with an expected decrease in delayed neutron yield at 14 MeV based on the well-established decrease in fission chain lengths with increasing energy of the neutron inducing fission.

Preliminary experimental data on absolute delayed neutron yields from 14 MeV fission of U^{238} and Th^{232} have been obtained by two different methods (modulated neutron beam and single pulse irradiations on the CW accelerator). Two different detectors (Hanson-McKibben "long counter," and He^3 proportional counter array -- cf. Figure 1) have been used in the course of these measurements. The experimental results summarized in Table

II indicate that U^{238} and Th^{232} delayed neutron yields do not increase with energy as previously reported in the literature, but remain essentially constant (or decrease somewhat) as expected, based on the behavior of fission mass and charge distributions with increasing neutron energy. It should be noted that the 14 MeV yield data in Table II are preliminary; conservatively-estimated experimental uncertainties are indicated. (Perturbations due to sample self-multiplication effects are not expected to be appreciable here.) These data are presently being refined and extended; also preparations are being made for absolute yield measurements on the other major fission species.

TABLE II

ABSOLUTE DELAYED NEUTRON YIELDS IN U^{238} AND Th^{232} FISSION

Fission Species	Absolute Delayed Neutron Yield: Fission Spectrum ($E_n \sim 3$ MeV)	$E_n = 14$ MeV
U^{238}	0.0412 ± 0.0017	0.036 ± 0.007
Th^{232}	0.0496 ± 0.0020	0.031 ± 0.009

DENSE PLASMA FOCUS (DPF) SOURCE

The existing N-6 Cockcroft-Walton accelerator provides an adequate pulsed neutron source for basic measurements necessary to the development of detection, identification and analysis techniques, and for nuclear safeguards research in general. However for the development of practical high-efficiency DIA systems, a more compact, more intense source of neutrons is required for a number of reasons. A comprehensive survey of existing and foreseeable sources has indicated that the pulsed neutron source which is best suited for DIA applications is the Dense Plasma Focus (DPF) device recently developed at Los Alamos (cf. J. W. Mather, "An Intense Source of neutrons from the Dense Plasma Focus," Session IVc, Symposium on Intense Neutron Sources, Santa Fe, N. M., September 19-23, 1966).

In the DPF device, stored magnetic energy is rapidly converted to plasma energy and then compressed by its self magnetic field. Plasma pressures (nkT) of the order of 3×10^5 atmospheres are developed in a region of ~0.5 mm diameter and 3.5 mm in length. The resulting neutron yield is $\sim 3 \times 10^{12}$ D, T neutrons in ~0.15 μsec, and is not inconsistent with the assumption of thermonuclear origin. Experimental evidence indicates that scaling of the DPF to larger input energies and larger physical sizes (but still relatively compact and mobile) should permit correspondingly higher neutron production e.g. scaling from present condenser energy of ~40 KJ to the 250 KJ range should result in a factor of ~5 increase in neutron yield.

Complete engineering and construction costs of the DPF source are included in the FY-1968 equipment budget of Group N-6, and engineering development and adaptation of the DPF pulsed neutron source to specific DIA requirements have already been initiated.

OTHER LASL ACTIVITIES IN SUPPORT OF NUCLEAR SAFEGUARDS RESEARCH

Radiochemical Analysis (J-11)

The total number of fissions in each sample irradiated at the CW accelerator (both beam modulation and single pulse irradiations) was determined by standard counting of the 67 hr. β-activity from Mo^{99} which was chemically separated from each irradiated sample. Measured β-yield is converted to number of fissions by an appropriate "K" factor determined from absolute fission counting measurements.

Source Radiography and Activation (GMX-1 and P-2)

The SbBe source used for detector response calibration at low energy was X-ray radiographed to determine its structural composition prior to thermal neutron activation in the "Water Boiler" thermal reactor, at Omega Site, LASL.

Neutron Source Calibration (P-6)

Four fast neutron sources were calibrated in the Los Alamos standard graphite pile. Calibrated source strengths (relative to PuBe standard source) were determined in the "Sigma pile" as follows:

Source	$\overline{E}_n$ (MeV)	Q (n/sec)
SbBe	~0.025	2.07×10^5 (1-30-67)
AmLi	~0.4	3.80×10^4
Cf^{252}	~2	4.16×10^5 (1-25-67)
PuBe	~4	6.43×10^5

Mass Spectrometric and Chemical Analysis (W-7 and CMB-1)

Depleted U^{238} samples were analyzed by mass spectrometry for uranium isotopic composition and by spectrochemical analysis for determination of trace concentrations of metal impurities. (No significant impurities found.)

PUBLICATIONS

1. "International Cooperation and the Control of Nuclear Energy," General Atomic Colloquium, invited talk, John J. Hopkins Laboratory, General Atomic, LaJolla, California, December 9, 1966.
2. "International Symposium on Intense Neutron Sources" (with P Division), Physics Today, 19 No. 12, 103 (1966).
3. "A Procedure for Calculating the Parameters Measured by Modified-Pulse-Source Experiments," Trans. Am. Nucl. Soc. 9 2 510 (1966).
4. "International Activities in Nuclear Inspection and Safeguards," banquet address, American Nuclear Society, National Topical Meeting on Coupled Reactor Kinetics, Texas A and M University, January 23-25, 1967.
5. "Delayed Fission Neutron Data in Reactor Physics and Design," abstract of paper to be presented at International Atomic Energy Agency, Panel on Delayed Fission Neutrons, Vienna, Austria, April 24-27, 1967.